

REMARKS

In response to the above Office Action, claim 1 has been amended to limit the lower amount of the monohalogenated hydrocarbon compound used in the claimed process to 0.625 moles. The claimed range in paragraph b. of claim 1 is therefore now 0.625 to 40 moles. As a result, claim 10 has been cancelled and claim 11 and 12 amended.

Support for this lower amount of the monohalogenated hydrocarbon, which is limited in the claim to n-butyl chloride, can be found in Example 2. In the Reply filed September 27, 2007, on page 4 it was noted that the molar ratio of the monohalogenated hydrocarbon compound to the transition metal of catalyst introduced into the reactor in Example 2 was "0.6." It was further noted that the ratio could be calculated using the transition metal content of the catalyst fed to the reactor and its introduction rate relative to the introduction rate of the monohalogenated hydrocarbon.

In Example 2, the titanium content of the catalyst was 4.3 weight %, and it was fed into the reactor at a rate of 25 g/h. The n-butyl chloride was fed at a rate of 14 mmol/h. See page 10, lines 21 and 30-32 of the specification.

Thus the amount of Ti fed to the reactor is $.043 \times 25 \text{ g/h} \times 1000 \text{ mg/1g}$ or 1075 mg of Ti per hour. The atomic weight of Ti is 47.9, so the amount in moles is $1075/47.9$ or 22.4 mmol Ti per hour. Thus the ratio of moles of n-butyl chloride to moles of transition metal is $14/22.4$ or more accurately 0.625.

Applicants' invention therefore, as now set forth in amended claim 1, relates to a process for the gas-phase (co-)polymerization of olefins in a fluidized bed reactor using a Ziegler-Natta type catalyst, said process comprising the addition into the reactor of an organoaluminium cocatalyst and of a monohalogenated hydrocarbon compound,

- a. wherein the molar ratio of the monohalogenated hydrocarbon compound to the cocatalyst is comprised between 0.02 and 0.2,
- b. wherein the monohalogenated hydrocarbon compound is added to the reactor in an amount comprised between 0.625 to 40 moles of monohalogenated hydrocarbon compound per mole of transition metal of catalyst introduced into the reactor, and
- c. wherein the monohalogenated hydrocarbon compound is n-butyl chloride.

Applicants found that this method of operating the reactor increases the level of polymerization activity of the Ziegler-Natta type catalyst during the (co-)polymerization of olefins, especially ethylene, and improves the processability of the polymer produced. See page 2, line 30 to page 3, line 18 of the specification. See also Examples 1-4 where n-butyl chloride was continuously fed to the reactor at a rate of from 14 to 39 mmol/h and at a molar ratio of from 0.05 to 0.15 of the chloride to the cocatalyst, which was triethylaluminium.

In the Office Action the Examiner continued to reject claims 1-13 under 35 U.S.C. § 103(a) for being obvious over U.S. Patent No. 5,990,251 to Gelus. As noted before, this reference belongs to the same assignee as the present application.

Gelus relates to a process for polymerizing olefin(s) in the presence of a Ziegler-Natta type catalyst and particularly to the use of halogenated hydrocarbon compounds as catalyst activating agents in an olefin polymerization process in the presence of a titanium-based catalyst of Ziegler-Natta type.

In column 2, lines 21-37 in Gelus, a large number of possible halogenated hydrocarbons are disclosed. In particular, the reference discloses that "the halogenated

hydrocarbon compound may be a chlorinated or brominated hydrocarbon. It may be a monohalogenated hydrocarbon, e.g., corresponding to the general formula R-X in which R denotes an alkyl group containing from 1 to 10, preferably from 1 to 4 carbon atoms, an aralkyl or aryl group containing from 6 to 14, preferably from 6 to 10 carbon atoms, and X denotes a halogen atom such as chlorine or bromine. The halogenated hydrocarbon compound may also be a polyhalogenated hydrocarbon, preferably containing 2 to 6, e.g., 2 to 4 halogen atoms such as chlorine or bromine, and 1 to 14, e.g., 1 to 4 carbon atoms per 1 molecule. Preferably it may also be a mono- or poly halogenated saturated hydrocarbon, such as the halogenated hydrocarbon compounds previously mentioned, e.g., methylene chloride, chloroform, carbon tetrachloride, trichloro-1, 1, 1 ethane or dichloro-1,2 ethane. Chloroform is employed most frequently."

As the Examiner notes, the reference does not explicitly mention "n-butyl chloride" as the monohalogenated hydrocarbon. Although it falls within the general formula R-X, relatively "light" halogenated hydrocarbons are preferred and all examples of Gelus use chloroform as the halogenated hydrocarbon compound.

The amount of halogenated hydrocarbon compound "advantageously" taught to be employed in Gelus is such that the molar ratio of the halogenated hydrocarbon to titanium introduced into the polymerization medium is "comprised between 0.001 and 0.15." Column 2, lines 42-47. The preferred ranges have an upper limit less than 0.1. Also as listed in the table below, all of the examples of Gelus have ratios (of chloroform to catalyst transition metal) below 0.1.

Ex/run	1	3B	3C	3D	4H	4I	4J	5	7	9
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Molar ratio 0.043 0.043 0.068 0.091 0.031 0.052 0.085 0.05 0.08 0.08

In addition, the graph of Fig. 2 of Gelus shows a molar ratio range of from 0.01 to 0.28 (points A to F).

In any event, it can be seen that the applicants' claimed molar ratio of 0.625 to 40 not only no longer overlaps that of Gelus, but is far away from it and far off the graph of Fig. 2 of Gelus.

Thus the present invention requires not only the selection of n-butyl chloride from all the other options for the halogenated hydrocarbons listed in Gelus, but also to then select a ratio of n-butyl chloride to Ti way above the disclosed ranges and exemplified values of Gelus. In fact, Gelus even teaches away from using significantly higher molar ratios of the present invention, since Comparative Examples 3E and 3F of Gelus clearly show the drawback of using higher ratios, i.e., 0.176 or 0.281 at least for chloroform. This is represented visually in the graph of Figure 2 in Gelus. Note how the activity of the catalyst significantly declines when ratios higher than about 0.10 of chloroform were used.

The Examiner believes that the selection of these two parameters, namely the use of n-butyl chloride and a molar ratio of 0.625 to 40 moles of n-butyl chloride per mole of transition metal of the catalyst, would somehow be obvious from the teachings of Gelus. However, it is submitted that these features would not be obvious in view of the explicit teachings in Gelus, particularly the preference for much lower ratios of the monohalogenated hydrocarbon to transition metal of the catalyst and the clear teaching in the Examples 1 and Fig 2 that any higher ratios of chloroform when used as the hydrocarbon are not advantageous. In fact, Gelus specifically teaches that:

"When the molar ratio of the quantity of the halogenated hydrocarbon compound to the quantity of titanium introduced into the polymerization medium is too high, it is found that the activity of the catalyst is not appreciably modified, or even is substantially reduced, particularly in a continuous polymerization process." (Emphasis added - column 3, lines 46-51).

Clearly the claimed range that starts at 0.625 would be considered "too high" by Gelus.

In support of applicants' invention, attached is a Declaration of one of the inventors, Claudine Lalanne-Magne, where the effect of a number of monohalogenated hydrocarbons, including chloroform and n-butyl chloride, on catalyst activity at a molar ratio of halogenated compound to Ti of 40:1 were compared. As noted, the catalyst was a Ziegler-Natta type catalyst similar to that used in the Examples of the application.

The results are shown in Figure 1 of the Declaration along with a reference sample in which no activity promoter was used. As noted in the Declaration with reference to Fig 1, "n-butylchloride (n-BuCl) provides a significant enhancement in activity (53%) compared to the unpromoted system."

It is to be further noted that the enhancement in activity at this ratio compared to chloroform, the only specific monohalogenated hydrocarbon actually used in Gelus, is over 80%! Moreover, as also noted in the Declaration, this level of activity for n-butyl chloride was 33% higher than the maximum increase taught by Gelus in Fig. 2 of the reference. N-butyl chloride also outperformed other monohalogenated hydrocarbons, i.e., ethyl chloride, n-propyl chloride, and n-pentyl chloride, all of which fall within the generic "R-X" compound of Gelus.

How then in view of this could it be said that a man skilled in the art would have specifically selected 1) n-butyl chloride and 2) at a molar ratio of 0.625 to 40 moles of n-butyl chloride per mole of transition metal of catalyst in the claimed process? As

noted in M.P.E.P. § 2144.05III, a prima facie case of obviousness may be rebutted by showing "that the art, in any material respect, teaches away from the claimed invention" and the Declaration clearly shows just that.

The Examiner notes in the Office Action on pager 5, lines 16-17 that she believes her rejection over Gelus to be justified on the grounds that Gelus "would have reasonably suggested to one having ordinary skill that n-butyl chloride would be as equally effective as chloroform." However, Mr. Lalanne-Magne's Declaration shows that it is not "equally effective," but in fact far superior to chloroform as well as other monohalogenated hydrocarbons of formula R-X. As noted in M.P.E.P. §716.02(a), results that are greater than those which would have been expected from the prior art are evidence of nonobviousness.

Withdrawal of Gelus as a ground of rejection under § 103(a) and allowance of claims 1-13 is therefore requested.

In view of the foregoing remarks, Applicants submit that this claimed invention, as amended, is neither anticipated nor rendered obvious in view of the prior art references cited against this application. Applicant therefore requests the entry of this Amendment, the Examiner's reconsideration and reexamination of the application, and the timely allowance of the pending claims.

Please grant any extensions of time required to enter this response and charge any additional required fees to Deposit Account 06-0916.

Respectfully submitted,

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Attachments: Declaration of Claudine Lalanne-Magne.

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